The clustering instability of inertial particles spatial distribution in turbulent flows

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A theory of clustering of inertial particles advected by a turbulent velocity field caused by an instability of their spatial distribution is suggested. The reason for the clustering instability is a combined effect of the particles inertia and a finite correlation time of the velocity field. The crucial parameter for the clustering instability is a size of the particles. The critical size is estimated for a strong clustering (with a finite fraction of particles in clusters) associated with the growth of the mean absolute value of the particles number density and for a weak clustering associated with the growth of the second and higher moments. A new concept of compressibility of the turbulent diffusion tensor caused by a finite correlation time of an incompressible velocity field is introduced. In this model of the velocity field, the field of Lagrangian trajectories is not divergence-free. A mechanism of saturation of the clustering instability associated with the particles collisions in the clusters is suggested. Applications of the analyzed effects to the dynamics of droplets in the turbulent atmosphere are discussed. An estimated nonlinear level of the saturation of the droplets number density in clouds exceeds by the orders of magnitude their mean number density. The critical size of cloud droplets required for clusters formation is more than 20μm.

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I. INTRODUCTION

Formation and evolution of aerosols and droplets inhomogeneities (clusters) are of fundamental significance in many areas of environmental sciences, physics of the atmosphere and meteorology (e.g., smog and fog formation, rain formation), transport and mixing in industrial turbulent flows (like spray drying, pulverized-coal-fired furnaces, cyclone dust separation, abrasive water-jet cutting) and in turbulent combustion (see, e.g., [1, 2, 3, 4, 5, 6, 7, 8]). The reason is that the direct, hydrodynamic, diffusional and thermal interactions of particles in dense clusters strongly affect the character of the involved phenomena. Thus, e.g., enhanced binary collisions between cloud droplets in dense clusters can cause fast broadening of droplet size spectrum and rain formation (see, e.g., [8]). Another example is combustion of pulverized coal or sprays whereby reaction rate of a single particle or a droplet differs considerably from a reaction rate of a coal particle or a droplet in a cluster (see, e.g., [9, 10]).

Analysis of experimental data shows that spatial distributions of droplets in clouds are strongly inhomogeneous (see, e.g., [11, 12, 13, 14]). Small-scale inhomogeneities in particles distribution were observed also in laboratory turbulent flows [15, 16, 17, 18].

It is well-known that turbulence results in a relaxation of inhomogeneities of concentration due to turbulent diffusion, whereas the opposite process, e.g., a preferential concentration (*clustering*) of droplets and particles in turbulent fluid flow still remains poorly understood.

In this paper we suggest a theory of clustering of particles and droplets in turbulent flows. The clusters of particles are formed due to an instability of their spatial distribution suggested in Ref. [19] and caused by a combined effect of a particle inertia and a finite velocity correlation time. Particles inside turbulent eddies are carried out to the boundary regions between them by inertial forces. This mechanism of the preferential concentration acts in all scales of turbulence, increasing toward small scales. An opposite process, a relaxation of clusters is caused by a scale-dependent turbulent diffusion. The turbulent diffusion decreases towards to smaller scales. Therefore, the clustering instability dominates in the Kolmogorov inner scale η, which separates inertial and viscous scales. Exponential growth of the number of particles in the clusters is saturated by their collisions.

In our previous study [19] we suggested and analyzed qualitatively an idea that inertia of particles may lead to their clustering. Later this idea was questioned by our quantitative analysis [20, 21] of the Kraichnan model of turbulent advection of particles by the delta-correlated in time random velocity field. It was proved that the clustering of inertial particles does not occur in the Kraichnan model. The latter result may be considered as counterexample.

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The main quantitative result of the theory of clustering instability of inertial particles, suggested in this paper, is *the existence of this instability* under some conditions that we determined. We showed that the inertia of the particles is only one of the *necessary conditions* for particles clustering in turbulent flow. In the present study we found a *second necessary condition* for the clustering instability: a finite correlation time of the fluid velocity field which in the suggested theory results in a nonzero divergence of the field of Lagrangian trajectories. This time is equal zero in the above mentioned Kraichnan model (see [22]) which was the reason for the disappearance of the instability in this particular model.

In this study we used a model of the turbulent velocity field with a finite correlation time which drastically changes the dynamics of inertial particles. In the framework of this model of the velocity field we rigorously derived the *sufficient conditions* for the clustering instability. We demonstrated the existence of the new phenomena of *strong* and *weak* clustering of inertial particles in a turbulent flow. These two types of the clustering instabilities have different physical meaning and different physical consequences in various phenomena. We computed also the instability thresholds which are different for the strong and weak clustering instabilities.

II. QUALITATIVE ANALYSIS OF STRONG AND WEAK CLUSTERING

A. Basic equations in the continuous media approximation

In this study we used the equation for the number density $n(t,\boldsymbol{r})$ of particles advected by a turbulent velocity field $\boldsymbol{u}(t,\boldsymbol{r})$:

$$\frac{\partial n(t,\boldsymbol{r})}{\partial t} + \boldsymbol{\nabla}\cdot[n(t,\boldsymbol{r})\,\boldsymbol{v}(t,\boldsymbol{r})] = D\,\Delta n(t,\boldsymbol{r})\,, \qquad (2.1)$$

where $D = kT/6\pi\nu\rho a$ is the coefficient of molecular (Brownian) diffusion, ν is the fluid kinematic viscosity, ρ and T are the fluid density and temperature, respectively, a is the radius of a particle and k is the Boltzmann constant. Due to inertia of particles their velocity $\boldsymbol{v}(t,\boldsymbol{r}) \neq \boldsymbol{u}(t,\boldsymbol{r})$, e.g., the field $\boldsymbol{v}(t,\boldsymbol{r})$ is not divergence-free even for $\operatorname{div}\boldsymbol{u} = 0$ (see [19]). Equation (2.1) implies conservation of the total number of particles in a closed volume. Consider

$$\Theta(t,\boldsymbol{r}) = n(t,\boldsymbol{r}) - \bar{n}\,, \qquad (2.2)$$

the deviation of $n(t,\boldsymbol{r})$ from the uniform mean number density of particles $\bar{n}$. Equation for $\Theta(t,\boldsymbol{r})$ follows from Eq. (2.1):

$$\begin{aligned}\frac{\partial\Theta(t,\boldsymbol{r})}{\partial t} &+ [\boldsymbol{v}(t,\boldsymbol{r})\cdot\boldsymbol{\nabla}]\Theta(t,\boldsymbol{r}) \qquad (2.3)\\ &= -\Theta(t,\boldsymbol{r})\operatorname{div}\boldsymbol{v}(t,\boldsymbol{r}) + D\Delta\Theta(t,\boldsymbol{r})\,.\end{aligned}$$

Here we assumed that the mean particles velocity is zero. We also neglected the term $\propto \bar{n}\operatorname{div}\boldsymbol{v}$ describing an effect of an external source of fluctuations. This term does not affect the growth rate of the instability. In the present study we investigate only the effect of self-excitation of the clustering instability, and we do not consider an effect of the source term on the dynamics of fluctuations. The source term $\propto \bar{n}\operatorname{div}\boldsymbol{v}$ causes another type of fluctuations of particle number density which are not related with an instability and are localized in the maximum scale of turbulent motions. A mechanism of these fluctuations is related with perturbations of the mean number density of particles by a random divergent velocity field. The magnitude of these fluctuations is much lower than that of fluctuations which are caused by the clustering instability.

In our qualitative analysis of the problem we use Eq. (2.3) written in a co-moving with a cluster reference frame. Formally, this may be done using the Belinicher-L'vov (BL) representation (for details, see [23, 24]). Let $\boldsymbol{\xi}_{\rm L}(t_0,\boldsymbol{r}|\,t)$ will be Lagrangian trajectory of the reference point [in the particle velocity field $\boldsymbol{v}(t,\boldsymbol{r})$] located at $\boldsymbol{r}$ at time t_0 and $\boldsymbol{\rho}_{\rm L}(t_0,\boldsymbol{r}|t)$ be an increment of the trajectory:

$$\begin{aligned}\boldsymbol{\rho}_{\rm L}(t_0,\boldsymbol{r}|t) &= \int_{t_0}^{t}\boldsymbol{v}[\tau,\boldsymbol{\xi}_{\rm L}(t_0,\boldsymbol{r}|\tau)]\,{\rm d}\tau\,, \qquad (2.4)\\ \boldsymbol{\xi}_{\rm L}(t_0,\boldsymbol{r}|t) &\equiv \boldsymbol{r} + \boldsymbol{\rho}_{\rm L}(t_0,\boldsymbol{r}|t)\,.\end{aligned}$$

By definition $\boldsymbol{\rho}_{\rm L}(t_0,\boldsymbol{r}|\,t_0) = 0$ and $\boldsymbol{\xi}(t_0,\boldsymbol{r}|\,t_0) = \boldsymbol{r}$. Define as $\boldsymbol{r}_0$ a position of a center of a cluster at the "initial" time $t_0 = 0$ (for the brevity of notations hereafter we skip the label t_0) and consider a "co-moving" reference frame with the position of the origin at $\boldsymbol{\zeta}_0(t) \equiv \boldsymbol{\zeta}(\boldsymbol{r}_0|\,t)$. Then BL velocity field $\tilde{\boldsymbol{v}}(\boldsymbol{r}_0|t,\boldsymbol{r})$ and BL velocity difference $\boldsymbol{W}(\boldsymbol{r}_0|t,\boldsymbol{r})$ are defined as

$$\begin{aligned}\tilde{\boldsymbol{v}}(\boldsymbol{r}_0|t,\boldsymbol{r}) &\equiv \boldsymbol{v}[t,\boldsymbol{r}+\boldsymbol{\rho}_{\rm L}(\boldsymbol{r}_0|t)]\,, \qquad (2.5)\\ \boldsymbol{W}(\boldsymbol{r}_0|t,\boldsymbol{r}) &\equiv \tilde{\boldsymbol{v}}(\boldsymbol{r}_0|t,\boldsymbol{r}) - \tilde{\boldsymbol{v}}(\boldsymbol{r}_0|t,\boldsymbol{r}_0)\,. \qquad (2.6)\end{aligned}$$

Actually the BL representation is very similar to the Lagrangian description of the velocity field. The difference between the two representations is that in the Lagrangian representation one follows the trajectory of every fluid particle $\boldsymbol{r} + \boldsymbol{\rho}_{\rm L}(\boldsymbol{r},t_0|t)$ (located at $\boldsymbol{r}$ at time $t = t_0$), whereas in the BL-representation there is a special initial point $\boldsymbol{r}_0$ (in our case the initial position of the center of the cluster) whose trajectory determines the new coordinate system (see [23, 24]). With time the BL-field $\tilde{\boldsymbol{v}}(\boldsymbol{r}_0|t,\boldsymbol{r})$ becomes very different from the Lagrangian velocity field. It must be noted that the simultaneous correlators of both, the Lagrangian and the BL-velocity fields, are identical to the simultaneous correlators of the Eulerian velocity $\boldsymbol{v}(\boldsymbol{r},t)$. The reason is that for stationary statistics the simultaneous correlators do not depend on t, and in particular one can assume $t = t_0$.

Similar to Eq. (2.5) let us introduce BL representation for $\Theta(t,\boldsymbol{r})$:

$$\tilde{\Theta}(\boldsymbol{r}_0|t,\boldsymbol{r}) \equiv \Theta[t,\boldsymbol{r}+\boldsymbol{\rho}_{\rm L}(\boldsymbol{r}_0|t)]\,. \qquad (2.7)$$

In BL variables defined by Eqs. (2.5)-(2.7), Eq. (2.3) reads:

$$\begin{aligned} &\frac{\partial\tilde\Theta(\boldsymbol{r}_0|t,\boldsymbol{r})}{\partial t}+[\boldsymbol{W}(\boldsymbol{r}_0|t,\boldsymbol{r})\cdot\boldsymbol{\nabla}]\tilde\Theta(\boldsymbol{r}_0|t,\boldsymbol{r}) \\ &= -\tilde\Theta(\boldsymbol{r}_0|t,\boldsymbol{r})\,\mathrm{div}\,\boldsymbol{W}(\boldsymbol{r}_0|t,\boldsymbol{r})+D\,\Delta\,\tilde\Theta(\boldsymbol{r}_0|t,\boldsymbol{r}) \ . \end{aligned} \tag{2.8}$$

The difference between Eqs. (2.3) and (2.8) is that Eq. (2.8) involves only velocity difference (2.6) in which the velocity $\tilde{\boldsymbol{v}}(\boldsymbol{r}_0|t,\boldsymbol{r}_0)$ of the cluster center is subtracted.

B. Rigid-cluster Approximation

Consider qualitatively a time evolution of different statistical moments

of the deviation $\Theta(t,\boldsymbol{r})$ defined by

$$\mathcal{M}_q(t)\equiv\langle|\Theta(t,\boldsymbol{r})|^q\rangle_v\,, \tag{2.9}$$

assuming that at the initial time, $t=0$, the spatial distribution of particles is almost homogeneous, all moments $\mathcal{M}_q(0)$ are small, where $\langle\cdot\rangle_v$ denotes the ensemble averaging over random velocity field $\boldsymbol{v}$. In order to eliminate the kinematic effect of sweeping of the cluster as a whole we consider Eq. (2.3) in the BL-representation, Eq. (2.8). Since the simultaneous moments of any field variables in the Eulerian and in the BL-representations coincide, the moments $\mathcal{M}_q(t)$ can be written as

$$\mathcal{M}_q(t)=\langle|\tilde\Theta(\boldsymbol{r}_0|t,\boldsymbol{r})|^q\rangle_v \ . \tag{2.10}$$

Our conjecture is that on a qualitative level we can consider the role of each term in the Eq. (2.8) separately, assuming some reasonable, time independent, *frozen* shape $\theta(x)$ of a distribution $\tilde\Theta(\boldsymbol{r}_0|t,\boldsymbol{r})$ inside a cluster:

$$\tilde\Theta(\boldsymbol{r}_0|t,\boldsymbol{r})=A(t)\,\theta\Big(\frac{|\boldsymbol{r}-\boldsymbol{r}_0|}{\ell_{\rm cl}}\Big) \ . \tag{2.11}$$

Here $A(t)$ is time-dependent amplitude of a cluster, $\ell_{\rm cl}$ is the characteristic width of the cluster. Shape function $\theta(x)$ may be chosen with a maximum equal one at $x=0$ and unit width. Real shapes of various clusters in the turbulent ensemble are determined by a competition of different terms in the evolution equation (2.8). However, we believe that particular shapes affect only numerical factors in the expression for the growth rate of clusters and do not effect their functional dependence on the parameters of the problem which is considered in this subsection.

1. *Effect of turbulent diffusion*

The advective term in the LHS of Eq. (2.8) results in turbulent diffusion inside the cluster. This effect may be modelled by renormalization of the molecular diffusion coefficient D in the right hand side (RHS) of Eq. (2.8) by the effective turbulent diffusion coefficient $D_{\rm T}$ with a usual estimate of $D_{\rm T}$:

$$D\to D+D_{\rm T}\,,\quad D_{\rm T}\sim\ell_{\rm cl}v_{\rm cl}/3 \ . \tag{2.12}$$

Hereafter $v_{\rm cl}$ is the mean square velocity of particles at the scale $\ell_{\rm cl}$. Instead of the full Eq. (2.8) consider now a model equation

$$\frac{\partial\tilde\Theta(\boldsymbol{r}_0|t,\boldsymbol{r})}{\partial t}=D_{\rm T}\,\Delta\,\tilde\Theta(\boldsymbol{r}_0|t,\boldsymbol{r})\,, \tag{2.13}$$

which accounts only for turbulent diffusion. Equations (2.10) and (2.13) yield:

$$\partial\mathcal{M}_q(t)/\partial t\simeq q\langle|\tilde\Theta(\boldsymbol{r}_0|t,\boldsymbol{r})|^{q-1}D_{\rm T}\Delta|\tilde\Theta(\boldsymbol{r}_0|t,\boldsymbol{r})|\rangle_w. \tag{2.14}$$

Substituting distribution (2.11) we estimate Laplacian in Eq. (2.14) as $-1/\ell_{\rm cl}^2$. Equations (2.13) and (2.14) imply that

$$\partial\mathcal{M}_q(t)/\partial t=-qD_{\rm T}\mathcal{M}_q(t)/\ell_{\rm cl}^2 \ . \tag{2.15}$$

The solution of Eq. (2.15) reads:

$$\begin{aligned} \mathcal{M}_q(t) &= \mathcal{M}_q(0)\exp[-\gamma_{\rm dif}(q)\,t]\,, \\ \gamma_{\rm dif}(q) &\sim qD_{\rm T}/\ell_{\rm cl}^2\,, \end{aligned} \tag{2.16}$$

where $\gamma_{\rm dif}(q)$ denotes a contribution to the damping rate of $\mathcal{M}_q(t)$ caused by turbulent diffusion.

2. *Effect of particles inertia*

In this subsection we show that the term $-\tilde\Theta\,\mathrm{div}\,\boldsymbol{W}$ in the RHS of Eq. (2.8) can result in an exponential growth of $\mathcal{M}_q(t)\propto\exp[\gamma_{\rm in}(q)\,t]$, i.e., in the instability. We denoted here the contribution to the growth rate of $\mathcal{M}_q(t)$, caused by the inertia of particles, by $\gamma_{\rm in}(q)$. In order to evaluate $\gamma_{\rm in}(q)$ we neglect now in Eq. (2.8) both, the convective term in the LHS of this equation (*i.e.* the the turbulent velocity difference inside the cluster) and the molecular diffusion term. The resulting equation reads:

$$\frac{\partial\tilde\Theta(\boldsymbol{r}_0|t,\boldsymbol{r})}{\partial t}=-\tilde\Theta(\boldsymbol{r}_0|t,\boldsymbol{r})\,\mathrm{div}\,\boldsymbol{W}(\boldsymbol{r}_0|t,\boldsymbol{r}) \ . \tag{2.17}$$

The main contribution to the BL-velocity difference $\boldsymbol{W}(\boldsymbol{r}_0|t,\boldsymbol{r})$ in the RHS of this equation is due to the eddies with size $\ell_{\rm cl}$, the characteristic size of the cluster. Denote by $v_{\rm cl}$ the characteristic velocity of these eddies and by $\tau_v\sim\ell_{\rm cl}/v_{\rm cl}$ the corresponding correlation time. In our qualitative analysis we neglect the $\boldsymbol{r}$ dependence of $\mathrm{div}\,\boldsymbol{W}(\boldsymbol{r}_0|t,\boldsymbol{r})$ inside the cluster and consider the divergence in Eq. (2.17) as a random process $b(t)$ with a correlation time τ_v:

$$\mathrm{div}\,\boldsymbol{W}(\boldsymbol{r}_0|t,\boldsymbol{r})\to b(t) \ . \tag{2.18}$$

Together with the decomposition (2.11) this yields the following equation for the cluster amplitude $A(t)$:

$$\frac{\partial A(t)}{\partial t}=-A(t)\,b(t) \ . \tag{2.19}$$

The solution of Eq. (2.19) reads:

$$A(t) = A_0 \, \exp[-I(t)] \, , \quad I(t) \equiv \int_0^t b(\tau) d\tau \ . \quad (2.20)$$

Integral $I(t)$ in Eq. (2.20) can be rewritten as a sum of integrals I_n over small time intervals τ_v:

$$I(t) = \sum_{n=1}^{t/\tau_v} I_n \, , \quad I_n(t) \equiv \int\limits_{(n-1)\tau_v}^{n\tau_v} b(\tau) \, d\tau \ .$$

In our qualitative analysis integrals I_n may be considered as independent random variables. Using the central limit theorem we estimate the total integral

$$I(t) \sim \sqrt{\langle I_n^2 \rangle_v} \sqrt{N} \zeta \, , \quad \langle I_n^2 \rangle_v = \langle b^2 \rangle_v \, \tau_v^2 \, ,$$

where $\langle \ldots \rangle_v$ denotes averaging over turbulent velocity ensemble, ζ is a Gaussian random variable with zero mean and unit variance, $N = t/\tau_v$. Now we calculate

$$\mathcal{M}_q(t) = \int \Theta^q P(\zeta) \, d\zeta \, , \quad P(\zeta) = (1/\sqrt{2\pi}) \exp(-\zeta^2/2) \ .$$

Therefore, $\mathcal{M}_q(t) = J_1 \exp(q^2 S^2 N/2)$, where

$$J_1 = (1/\sqrt{2\pi}) \int \exp[-(\zeta - qS\sqrt{N})^2/2] \, d\zeta \sim 1 \ .$$

Since the main contribution to the integral J_1 arises from $\zeta \sim qS\sqrt{N}$, the parameter q cannot be large. In this approximation the $q-$moments

$$\mathcal{M}_q(t) = \mathcal{M}_q(0) \exp[\gamma_{\rm in}(q) t]$$

with $\gamma_{\rm in}(q)$ being the growth rate of the q-th moment due to particles inertia which is given by

$$\gamma_{\rm in}(q) \sim \frac{1}{2} \langle \tau_v [\operatorname{div} \boldsymbol{W}(\boldsymbol{r}_0|t, \boldsymbol{r})]^2 \rangle_v q^2 \ . \qquad (2.21)$$

3. *Qualitative picture of the clustering instability*

In previous subsections we evaluated the contributions to the growth rate of $\mathcal{M}_q(t)$ due to the turbulent diffusion $\gamma_{\rm dif}(q)$, Eq. (2.16), and due to the particles inertia $\gamma_{\rm in}(q)$, Eq. (2.21). The total growth rate may be evaluated as a sum of these contributions:

$$\begin{aligned} \mathcal{M}_q(t) &= \mathcal{M}_q(0) \exp(\gamma_q t) \, , \qquad (2.22) \\ \gamma_q &\simeq \gamma_{\rm dif}(q) + \gamma_{\rm in}(q) \, , \\ \gamma_q &\sim -q D_{\rm T}/\ell_{\rm cl} + \frac{1}{2} \langle \tau_v [\operatorname{div} \boldsymbol{W}(\boldsymbol{r}_0|t, \boldsymbol{r})]^2 \rangle_v q^2 \ . \end{aligned}$$

Clearly, the instability is caused by a nonzero value of $\langle \tau_v (\operatorname{div} \boldsymbol{W})^2 \rangle$, *i.e.,* by a *compressibility of the particle velocity field* $\boldsymbol{v}(t, \boldsymbol{r})$.

Compressibility of fluid velocity itself $\boldsymbol{u}(t, \boldsymbol{r})$ (including atmospheric turbulence) is often negligible, i.e., $\operatorname{div} \boldsymbol{u} \approx 0$. However, due to the effect of particles inertia their velocity $\boldsymbol{v}(t, \boldsymbol{r})$ does not coincide with $\boldsymbol{u}(t, \boldsymbol{r})$ (see, e.g., [25, 26, 27, 28]), and a degree of compressibility, σ_v, of the field $\boldsymbol{v}(t, \boldsymbol{r})$, may be of the order of unity[19, 20, 29].

Parameter σ_v is defined as

$$\sigma_v \equiv \langle [\operatorname{div} \boldsymbol{v}]^2 \rangle / \langle |\boldsymbol{\nabla} \times \boldsymbol{v}|^2 \rangle \ . \qquad (2.23)$$

Note that parameter σ_v is independent of the scale of the turbulent velocity field. It characterizes a compressible part of the velocity field as a whole. The main contribution to this parameter comes from the scales which are of the order of the Kolmogorov scale η.

For inertial particles $\operatorname{div} \boldsymbol{v} \sim \tau_{\rm p} \Delta P/\rho$, where $\tau_{\rm p}$ is the particle response time,

$$\tau_{\rm p} = m_{\rm p}/6\pi\rho\nu a = 2\rho_{\rm p} a^2/9\rho\nu \, , \qquad (2.24)$$

where $m_{\rm p}$ and $\rho_{\rm p}$ are the mass and material density of particles, respectively. The fluid flow parameters are: pressure P, Reynolds number $\mathcal{R}e = L u_{\rm T}/\nu$, the dissipative scale of turbulence $\eta = L \, \mathcal{R}e^{-3/4}$, the maximum scale of turbulent motions L and the turbulent velocity $u_{\rm T}$ in the scale L. Now we can estimate σ_v as

$$\sigma_v \simeq (\rho_{\rm p}/\rho)^2 (a/\eta)^4 \equiv (a/a_*)^4 \qquad (2.25)$$

(see [20]), where a_* is a characteristic radius of particles. For $a > a_*$ it is plausible to correct this estimate as follows:

$$\sigma_v \sim \frac{a^4}{a^4 + a_*^4} \ . \qquad (2.26)$$

For water droplets in the atmosphere $\rho_{\rm p}/\rho \simeq 10^3$ and $a_* \simeq \eta/30$. For the typical value of $\eta \simeq 1$mm it yields $a_* \simeq 30\mu$m. On windy days when η decreases, the value of a_* correspondingly becomes smaller.

Then we estimate $\langle \tau_v [\operatorname{div} \boldsymbol{v}]^2 \rangle$ as $2\sigma_v/\tau_v$, because $\langle [\operatorname{rot} \boldsymbol{v}]^2 \rangle \sim 2\tau_v^{-2}$. Assuming that the cluster size $\ell_{\rm cl}$ is of the order of the inner scale of turbulence, η, we have to identify τ_v with a turnover time of eddies in the inner scale η, $\tau_v \to \tau_\eta \equiv \eta/v_\eta = (L/u_{\rm T}) \, \mathcal{R}e^{-1/2}$. Thus, the growth rate γ_q of the q-th moment in Eq. (2.22) may be evaluated as

$$\gamma_q \simeq \gamma_{\rm cl} \, q(q - q_{\rm cr}) \, , \quad \gamma_{\rm cl} \sim \frac{\sigma_v}{\tau_\eta} \, , \quad q_{\rm cr} \sim \frac{1}{3\sigma_v} \ . \ (2.27)$$

Clearly, the moments with $q > q_{\rm cr}$ are unstable. Equations (2.25) and (2.27) imply that it happens when $a > a_{q,\rm cr}$ where $a_{q,\rm cr} = a_{1,\rm cr}/q^{1/4}$ is the value of a at which $q_{\rm cr} = q$. The largest value of $a_{q,\rm cr}$ corresponds to the instability of the first moment, $\langle |\Theta| \rangle$: $a_{1,\rm cr} \sim 0.8 \, a_*$, $a_{2,\rm cr} \approx 0.84 \, a_{1,\rm cr}$, $a_{3,\rm cr} \approx 0.76 \, a_{1,\rm cr}$, $a_{4\rm cr} \approx 0.71 \, a_{1,\rm cr}$, *etc.*

Note that if $\langle |\Theta| \rangle$ grows in time then almost all particles can be accumulated inside the clusters (if we neglect a nonlinear saturation of such growth). We define this case as a *strong clustering.* On the other hand, if $q_{\rm cr} > 1$ the first moment $\langle |\Theta| \rangle$ does not grow, and the clusters

contain a small fraction of the total number of particles. This does not mean that the instability of higher moments is not important. Thus, e.g., the rate of binary particles collisions is proportional to the square of their number density $\langle n^2\rangle = (\bar{n})^2 + \langle|\Theta|^2\rangle$. Therefore, the growth of the 2nd moment, $\langle|\Theta|^2\rangle$, (which we define as a *weak clustering*) results in that binary collisions occur mainly between particles inside the cluster. The latter can be important in coagulation of droplets in atmospheric clouds whereby the collisions between droplets play a crucial role in a rain formation. The growth of the q-th moment, $\langle|\Theta|^q\rangle$, results in that q-particles collisions occur mainly between particles inside the cluster. The growth of the negative moments of particles number density (possibly associated with formation of voids and cellular structures) was discussed in [30] (see also [31, 32]).

In the above qualitative analysis whereby we considered only one-point correlation functions of the number density of particles, we missed an important effect of an effective drift velocity which decreases a growth rate of the clustering instability. For the one-point correlation functions of the number density of particles the effective drift velocity is zero for homogeneous and isotropic turbulence. However, in the equations for two-point and multi-point correlation functions of the number density of particles the effective drift velocity is not zero and as we will see in the next section it increases a threshold for the clustering instability.

III. THE CLUSTERING INSTABILITY OF THE 2ND MOMENT

A. Basic equations

In the previous section we estimated the growth rates of all moments $\langle|\Theta|^q\rangle$. Here we present the results of a rigorous analysis of the evolution of the two-point 2nd moment

$$\Phi(t,\boldsymbol{R}) \equiv \langle\Theta(t,\boldsymbol{r})\Theta(t,\boldsymbol{r}+\boldsymbol{R})\rangle \ . \tag{3.1}$$

In this analysis we used stochastic calculus [e.g., Wiener path integral representation of the solution of the Cauchy problem for Eq. (2.1), Feynman-Kac formula and Cameron-Martin-Girsanov theorem]. The comprehensive description of this approach can be found in [21, 33, 34, 35].

We showed that a finite correlation time of a turbulent velocity plays a crucial role for the clustering instability. Notably, an equation for the second moment $\Phi(t,\boldsymbol{R})$ of the number density of inertial particles comprises spatial derivatives of high orders due to a non-local nature of turbulent transport of inertial particles in a random velocity field with a finite correlation time (see Appendix A and [20]). However, we found that equation for $\Phi(t,\boldsymbol{R})$ is a second-order partial differential equation at least for two models of a random velocity field:

*M*odel I. The random velocity with Gaussian statistics of the integrals $\int_0^t \boldsymbol{v}(t',\boldsymbol{\xi})\,dt'$ and $\int_0^t b(t',\boldsymbol{\xi})\,dt'$, see Appendix B.

*M*odel II. The Gaussian velocity field with a small yet finite correlation time, see Appendix C.

In both models equation for $\Phi(t,\boldsymbol{R})$ has the same form:

$$\begin{aligned}&\partial\Phi/\partial t = \hat{\mathcal{L}}\,\Phi(t,\boldsymbol{R}) \,, \\ &\hat{\mathcal{L}} = B(\boldsymbol{R}) + 2\boldsymbol{U}(\boldsymbol{R})\cdot\boldsymbol{\nabla} + \hat{D}_{\alpha\beta}(\boldsymbol{R})\nabla_\alpha\nabla_\beta \,,\end{aligned} \tag{3.2}$$

but with different expressions for its coefficients. The meaning of the coefficients $B(\boldsymbol{R})$, $\boldsymbol{U}(\boldsymbol{R})$ and $\hat{D}_{\alpha\beta}(\boldsymbol{R})$ is as follows:

– Function $B(\boldsymbol{R})$ is determined only by a compressibility of the velocity field and it causes generation of fluctuations of the number density of inertial particles.

– The vector $\boldsymbol{U}(\boldsymbol{R})$ determines a scale-dependent drift velocity which describes a transfer of fluctuations of the number density of inertial particles over the spectrum. Note that $\mathbf{U}(\boldsymbol{R}=0)=0$ whereas $B(\boldsymbol{R}=0)\neq 0$. For incompressible velocity field $\mathbf{U}(\boldsymbol{R})=0$, $B(\boldsymbol{R})=0$.

– The scale-dependent tensor of turbulent diffusion $\hat{D}_{\alpha\beta}(\boldsymbol{R})$ is also affected by the compressibility.

In very small scales this tensor is equal to the tensor of the molecular (Brownian) diffusion, while in the vicinity of the maximum scale of turbulent motions this tensor coincides with the usual tensor of turbulent diffusion. Tensor $\hat{D}_{\alpha\beta}(\boldsymbol{R})$ may be written as

$$\begin{aligned}\hat{D}_{\alpha\beta}(\boldsymbol{R}) &= 2D\delta_{\alpha\beta} + D^{\rm T}_{\alpha\beta}(\boldsymbol{R}) \,, \\ D^{\rm T}_{\alpha\beta}(\boldsymbol{R}) &= \tilde{D}^{\rm T}_{\alpha\beta}(0) - \tilde{D}^{\rm T}_{\alpha\beta}(\boldsymbol{R}) \ .\end{aligned} \tag{3.3}$$

In Appendix B we found that for Model I:

$$\begin{aligned}B(\boldsymbol{R}) &\approx 2\int_0^\infty \langle b[0,\boldsymbol{\xi}(\boldsymbol{r}_1|0)]b[\tau,\boldsymbol{\xi}(\boldsymbol{r}_2|\tau)]\rangle\,d\tau \,, \\ \boldsymbol{U}(\boldsymbol{R}) &\approx -2\int_0^\infty \langle \boldsymbol{v}[0,\boldsymbol{\xi}(\boldsymbol{r}_1|0)]b[\tau,\boldsymbol{\xi}(\boldsymbol{r}_2|\tau)]\rangle\,d\tau \,, \\ \tilde{D}^{\rm T}_{\alpha\beta}(\boldsymbol{R}) &\approx 2\int_0^\infty \langle v_\alpha[0,\boldsymbol{\xi}(\boldsymbol{r}_1|0)]v_\beta[\tau,\boldsymbol{\xi}(\boldsymbol{r}_2|\tau)]\rangle\,d\tau \,.\end{aligned} \tag{3.4}$$

For the δ-correlated in time random Gaussian compressible velocity field the operator $\hat{\mathcal{L}}$ is replaced by $\hat{\mathcal{L}}_0$ in the equation for the second moment $\Phi(t,\boldsymbol{R})$, where

$$\begin{aligned}&\hat{\mathcal{L}}_0 \equiv B_0(\boldsymbol{R}) + 2\boldsymbol{U}_0(\boldsymbol{R})\cdot\boldsymbol{\nabla} + \hat{D}_{\alpha\beta}(\boldsymbol{R})\nabla_\alpha\nabla_\beta \,, \\ &B_0(\boldsymbol{R}) = \nabla_\alpha\nabla_\beta\hat{D}_{\alpha\beta}(\boldsymbol{R}) \,, \\ &U_{0,\alpha}(\boldsymbol{R}) = \nabla_\beta\hat{D}_{\alpha\beta}(\boldsymbol{R})\end{aligned} \tag{3.5}$$

(for details see [20, 21]). In the δ-correlated in time velocity field the second moment $\Phi(t,\boldsymbol{R})$ can only decay in spite of the compressibility of the velocity field. The reason is that the differential operator $\hat{\mathcal{L}}_0 \equiv \nabla_\alpha\nabla_\beta\hat{D}_{\alpha\beta}(\boldsymbol{R})$ is adjoint to the operator $\hat{\mathcal{L}}_0^\dagger \equiv \hat{D}_{\alpha\beta}(\boldsymbol{R})\nabla_\alpha\nabla_\beta$ and their eigenvalues are equal. The damping rate for the equation

$$\partial\Phi/\partial t = \hat{\mathcal{L}}_0^\dagger\,\Phi(t,\boldsymbol{R}) \tag{3.6}$$

has been found in Ref. [36] for a compressible isotropic homogeneous turbulence in a dissipative range:

$$\gamma_2 = -\frac{(3-\sigma_{\rm T})^2}{6\,\tau_\eta(1+\sigma_{\rm T})(1+3\sigma_{\rm T})} \; . \tag{3.7}$$

Here $\sigma_{\rm T}$ is the degree of compressibility of the tensor $D^{\rm T}_{\alpha\beta}(\boldsymbol{R})$. For the δ-correlated in time incompressible velocity field ($\sigma_{\rm T}=0$) Eq. (3.6) was derived in Ref. [22]. Thus, for the Kraichnan model of turbulent advection (with a delta correlated in time velocity field) the clustering instability of the 2nd moment does not occur.

A general form of the turbulent diffusion tensor in a dissipative range is given by

$$\begin{aligned} & D^{\rm T}_{\alpha\beta}(\boldsymbol{R}) = (C_1 R^2 \delta_{\alpha\beta} + C_2 R_\alpha R_\beta)/\tau_\eta \,, \\ & C_1 = 2(2+\sigma_{\rm T})/3\,(1+\sigma_{\rm T}) \,, \\ & C_2 = 2(2\sigma_{\rm T}-1)/3\,(1+\sigma_{\rm T}) \; . \end{aligned} \tag{3.8}$$

The parameter $\sigma_{\rm T}$ is defined by analogy with Eq. (2.23):

$$\sigma_{\rm T} \equiv \frac{\boldsymbol{\nabla}\cdot \boldsymbol{D}_{\rm T}\cdot\boldsymbol{\nabla}}{\boldsymbol{\nabla}\times\boldsymbol{D}_{\rm T}\times\boldsymbol{\nabla}} = \frac{\nabla_\alpha\nabla_\beta D^{\rm T}_{\alpha\beta}(\boldsymbol{R})}{\nabla_\alpha\nabla_\beta D^{\rm T}_{\alpha'\beta'}(\boldsymbol{R})\epsilon_{\alpha\alpha'\gamma}\epsilon_{\beta\beta'\gamma}} \,, \tag{3.9}$$

where $\epsilon_{\alpha\beta\gamma}$ is the fully antisymmetric unit tensor. Equations (2.23) and (3.9) imply that $\sigma_{\rm T}=\sigma_v$ in the case of δ-correlated in time compressible velocity field. Equations (3.4) show that for a finite correlation time identities (3.5) are violated and

$$B(\boldsymbol{R}) \neq B_0(\boldsymbol{R}) \,, \quad \boldsymbol{U}(\boldsymbol{R}) \neq \boldsymbol{U}_0(\boldsymbol{R}) \; .$$

For a random incompressible velocity field with a finite correlation time the tensor of turbulent diffusion $D_{\alpha\beta}(\boldsymbol{R}) = \tau^{-1}\langle\xi_\alpha(\boldsymbol{r}_1)\xi_\beta(\boldsymbol{r}_2)\rangle$ [see Eq. (C5)] and the degree of compressibility of this tensor is

$$\sigma_{\rm T} = \frac{\langle(\boldsymbol{\nabla}\cdot\boldsymbol{\xi})^2\rangle}{\langle(\boldsymbol{\nabla}\times\boldsymbol{\xi})^2\rangle} \,, \tag{3.10}$$

where $\boldsymbol{\xi}(\boldsymbol{r}_1|t)$ is the Lagrangian displacement of a particle trajectory which paths through point $\boldsymbol{r}_1$ at $t=0$. Remarkably that Taylor [37] obtained the coefficient of turbulent diffusion for the mean field in the form $D_{\rm T}(\boldsymbol{R}=0) = \tau^{-1}\langle\xi_\alpha(\boldsymbol{r}_1|t)\xi_\alpha(\boldsymbol{r}_1|t)\rangle$.

B. Clustering instability in Model I

Let us study the clustering instability for the model of the random velocity with Gaussian statistics of the integrals

$$\int_0^t \boldsymbol{v}(t',\boldsymbol{\xi})\,dt' \,, \quad \int_0^t b(t',\boldsymbol{\xi})\,dt' \,,$$

see Appendix B. In this model Eq. (3.2) in a non-dimensional form reads:

$$\begin{aligned} \frac{\partial\Phi}{\partial\tilde t} &= \frac{\Phi''}{m(r)} + \Big[\frac{1}{m(r)} + (U-C_2)r^2\Big]\frac{2\,\Phi'}{r} + B\Phi \,, \\ 1/m(r) &\equiv (C_1+C_2)r^2 + 2/{\rm Sc} \,, \end{aligned} \tag{3.11}$$

where $\boldsymbol{U}\equiv U\boldsymbol{R}$ and Sc$=\nu/D$ is the Schmidt number. For small inertial particles advected by air flow Sc$\gg 1$. The non-dimensional variables in Eq. (3.11) are $r\equiv R/\eta$ and $\tilde t = t/\tau_\eta$, B and U are measured in the units τ_η^{-1}. Consider a solution of Eq. (3.11) in two spatial regions.

a. *Molecular diffusion region of scales.* In this region $r \ll {\rm Sc}^{-1/2}$, and all terms $\propto r^2$ (with C_1, C_2 and U) may be neglected. Then the solution of Eq. (3.11) is given by

$$\Phi(r) = (1-\alpha r^2)\exp(\gamma_2 t) \,,$$

where

$$\alpha \;=\; {\rm Sc}(B-\gamma_2\tau_\eta)/12 \,, \quad B > \gamma_2\tau_\eta \; .$$

b. *Turbulent diffusion region of scales.* In this region ${\rm Sc}^{-1/2} \ll r \ll 1$, the molecular diffusion term $\propto 1/{\rm Sc}$ is negligible. Thus, the solution of (3.11) in this region is

$$\Phi(r) = A_1 r^{-\lambda}\exp(\gamma_2 t) \,,$$

where

$$\begin{aligned} \lambda &= (C_1 - C_2 + 2U \pm iC_3)/2(C_1+C_2) \,, \\ C_3^2 &= 4(B-\gamma_2\tau_\eta)(C_1+C_2) - (C_1-C_2+2U)^2 \; . \end{aligned}$$

Since the total number of particles in a closed volume is conserved:

$$\int_0^\infty r^2\Phi(r)\,dr = 0 \; .$$

This implies that $C_3^2 > 0$, and therefore λ is a complex number. Since the correlation function $\Phi(r)$ has a global maximum at $r=0$, $C_1 > C_2 - 2U$. The latter condition for very small U yields $\sigma_T \leq 3$. For $r \gg 1$ the solution for $\Phi(r)$ decays sharply with r. The growth rate γ_2 of the second moment of particles number density can be obtained by matching the correlation function $\Phi(r)$ and its first derivative $\Phi'(r)$ at the boundaries of the above regions, i.e., at the points $r={\rm Sc}^{-1/2}$ and $r=1$. The matching yields $C_3/2(C_1+C_2) \approx 2\pi/\ln{\rm Sc}$. Thus,

$$\begin{aligned} \gamma_2 \;=\; & \frac{1}{\tau_\eta(1+3\sigma_{\rm T})}\Big[\frac{200\sigma_{\rm U}(\sigma_{\rm T}-\sigma_{\rm U})}{3(1+\sigma_{\rm U})} - \frac{(3-\sigma_{\rm T})^2}{6(1+\sigma_{\rm T})} \\ & - \frac{3\pi^2(1+3\sigma_{\rm T})^2}{(1+\sigma_{\rm T})\ln^2{\rm Sc}}\Big] + \frac{20(\sigma_{\rm B}-\sigma_{\rm U})}{\tau_\eta(1+\sigma_{\rm B})(1+\sigma_{\rm U})} \,, \end{aligned} \tag{3.12}$$

where we introduced parameters $\sigma_{\rm B}$ and $\sigma_{\rm U}$ defined by

$$B = 20\sigma_{\rm B}/(1+\sigma_{\rm B}) \,, \quad U = 20\sigma_{\rm U}/3(1+\sigma_{\rm U}) \; . \tag{3.13}$$

Note that the parameters $\sigma_{\rm B} \approx \sigma_{\rm U} \sim \sigma_v$. For the δ-correlated in time random compressible velocity field $\sigma_{\rm B}=\sigma_{\rm U}=\sigma_{\rm T}=\sigma_v$. Figure 1 shows the range of parameters $(\sigma_v, \sigma_{\rm T})$ for $\sigma_{\rm B}=\sigma_{\rm U}=\sigma_v$ in the case of Sc $=10^3$ (curve c), Sc $=10^5$ (curve b) and Sc $\to\infty$ (curve a). The dashed line $\sigma_v=\sigma_{\rm T}$ corresponds to the δ-correlated in time random compressible velocity field. This is a limiting line for the curve "a". Figure 1 demonstrates

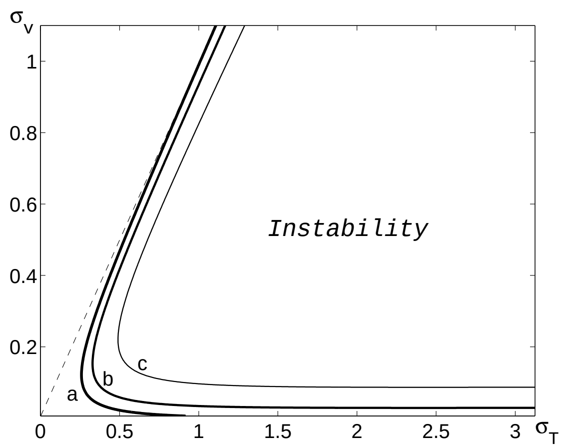

FIG. 1: The range of parameters $(\sigma_v, \sigma_{\rm T})$ for $\sigma_{\rm B} = \sigma_{\rm U} = \sigma_v$ in the case of $\mathrm{Sc} = 10^3$ (curve c), $\mathrm{Sc} = 10^5$ (curve b) and $\mathrm{Sc} \to \infty$ (curve a). The dashed line $\sigma_v = \sigma_{\rm T}$ corresponds to the δ-correlated in time random compressible velocity field.

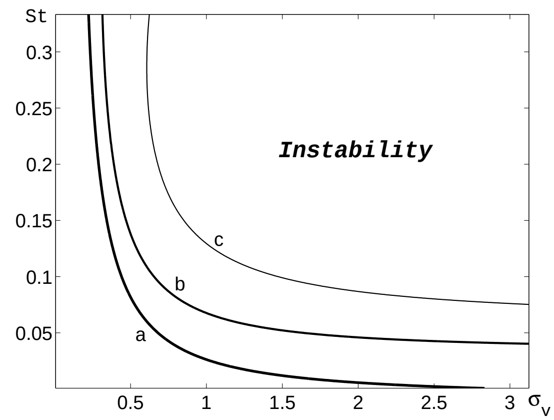

FIG. 2: The range of parameters (Sc, σ_v) for $\mathrm{Sc} = 10^3$ (curve c), $\mathrm{Sc} = 10^5$ (curve b) and $\mathrm{Sc} \to \infty$ (curve a). The line $\mathrm{Sc} = 0$ corresponds to the δ-correlated in time random compressible velocity field.

that even a very small deviations from the δ-correlated in time random compressible velocity field results in the instability of the second moment of the number density of inertial particles. The minimum value of $\sigma_{\rm T}$ required for the clustering instability is $\sigma_{\rm T} \approx 0.26$ and a corresponding value of $\sigma_v \approx 0.12$ (see Fig. 1). For smaller value of σ_v the clustering instability can occur, but it requires larger values of $\sigma_{\rm T}$.

Notably, in Model II of a random velocity field (i.e., the Gaussian velocity field with a small yet finite correlation time) the clustering instability occurs when $\sigma_v > 0.2$ (see Appendix C). Indeed, the growth rate γ_2 of the second moment of particles number density is determined by equation:

$$
\begin{aligned}
\Gamma &= \tilde B(\sigma_v)\mathrm{St}^2 - \frac{(3-\sigma_v)^2}{6(1+\sigma_v)(1+3\sigma_v)} \\
&\quad - \frac{8(1+3\sigma_v)}{3(1+\sigma_v)}\left(\frac{\pi}{\ln \mathrm{Sc}}\right)^2 , \\
\tilde B(\sigma_v) &= 12\left(b_2 + \frac{b_3 a_1}{4a_2^2} - \frac{b_1}{2a_2}\right) ,
\end{aligned}
$$

where St$= \bar\tau_{\rm ren}/\tau_\eta$ is the Strouhal number, $\Gamma = \gamma_2(1 + \bar\tau_{\rm ren}\gamma_2)^2$, and

$$
\begin{aligned}
a_1 &= \frac{2(19\sigma_v + 3)}{3(1+\sigma_v)} , \qquad a_2 = \frac{2(3\sigma_v + 1)}{3(1+\sigma_v)} , \\
b_1 &= -\frac{1}{27(1+\sigma_v)^2}(12 - 1278\sigma_v - 3067\sigma_v^2) , \\
b_2 &= \frac{850}{9}\left(\frac{\sigma_v}{1+\sigma_v}\right)^2 , \\
b_3 &= \frac{1}{27(1+\sigma_v)^2}(36 + 466\sigma_v + 2499\sigma_v^2) .
\end{aligned}
$$

Figure 2 shows the range of parameters (Sc, σ_v) for $\mathrm{Sc} = 10^3$ (curve c), $\mathrm{Sc} = 10^5$ (curve b) and $\mathrm{Sc} \to \infty$ (curve a).

It is seen from FIG. 2 that for $\sigma_v > 0.2$ the second-order correlation function of the number density of inertial particles can grow in time exponentially (i.e., $\gamma_2 > 0$) even for very small Strouhal numbers. For example, in the vicinity of $\sigma_v = 3$, the growth rate γ_2 of the clustering instability of the second-order correlation function is given by

$$
\gamma_2 = 4\times 10^3\left(\frac{\tau u_\eta}{\eta}\right)^2 - \frac{(3-\sigma_v)^2}{240} - \frac{20}{3}\left(\frac{\pi}{\ln \mathrm{Sc}}\right)^2 . \quad (3.14)
$$

The sufficient condition for the exponential growth of the second moment of a number density of inertial particles is $\mathrm{Sc} > \mathrm{Sc}^{(\rm cr)}$, where the critical Schmidt number $\mathrm{Sc}^{(\rm cr)}$ is given by $\mathrm{Sc}^{(\rm cr)} = \mathrm{Sc}(\gamma_2 = 0)$. The clustering instability occurs when the degree of compressibility of particles velocity $\sigma_v > 0.2$, i.e., for particles and droplets with the radius $a_* > 25.4\,\mu$m. Equation (2.27) also yields a similar value $\sigma_{\rm cr} \sim 1/6$ for the threshold of the instability of the 2nd moment (at $q_{\rm cr} = 2$). Note that Eq. (3.7) is written for $\mathrm{Sc} \to \infty$.

IV. NONLINEAR EFFECTS

The compressibility of the turbulent velocity field with a finite correlation time can cause the exponential growth of the moments of particles number density. This small-scale instability results in formation of strong inhomogeneities (clusters) in particles spatial distributions. The linear analysis does not allow to determine a mechanism of saturation of the clustering instability. As can be seen from Eq. (3.12) molecular diffusion only depletes the growth rates of the clustering instability at the linear stage (contrary to the instability discussed in Ref.

[30]). The clustering instability is saturated by nonlinear effects.

Now let us discuss a mechanism of the nonlinear saturation of the clustering instability using on the example of atmospheric turbulence with characteristic parameters: $\eta \sim 1$mm, $\tau_\eta \sim (0.1 - 0.01)$s. A momentum coupling of particles and turbulent fluid is essential when $m_{\rm p} n_{\rm cl} \sim \rho$, i.e., the mass loading parameter $\phi = m_{\rm p} n_{\rm cl}/\rho$ is of the order of unity (see, e.g., [1]). This condition implies that the kinetic energy of fluid $\rho \langle \boldsymbol{u}^2 \rangle$ is of the order of the particles kinetic energy $m_{\rm p} n_{\rm cl} \langle \boldsymbol{v}^2 \rangle$, where $|\boldsymbol{u}| \sim |\boldsymbol{v}|$. This yields:

$$n_{\rm cl} \sim a^{-3} (\rho / 3\rho_{\rm p}) \; . \tag{4.1}$$

For water droplets $\rho_{\rm p}/\rho \sim 10^3$. Thus, for $a = a_* \sim 30\mu$m we obtain $n_{\rm cl} \sim 10^4$ cm^{-3} and the total number of particles in the cluster of size η, $N_{\rm cl} \simeq \eta^3 n_{\rm cl} \sim 10$. This values may be considered as a lower estimate for the "two-way coupling" when the effect of fluid on particles has to be considered together with the feed-back effect of the particles on the carrier fluid. However, it was found in [38] that turbulence modification by particles is governed by the ratio of the particle energy and *the total energy of the suspension* (rather then the energy of the carrier fluid) and thus by parameter $\phi(1+\phi)$ (rather then by ϕ itself). Thus we expect that the two-way coupling can only mitigate but not stop the clustering instability.

An actual mechanism of the nonlinear saturation of the clustering instability is "four way coupling" when the particle-particle interaction is also important. In this situation the particles collisions result in effective particle pressure which prevents further grows of concentration. Particles collisions play essential role when during the life-time of a cluster the total number of collisions is of the order of number of particles in the cluster. The rate of collisions $J \sim n_{\rm cl}/\tau_\eta$ can be estimated as $J \sim 4\pi a^2 n_{\rm cl}^2 |\boldsymbol{v}_{\rm rel}|$. The relative velocity $\boldsymbol{v}_{\rm rel}$ of colliding particles with different but comparable sizes can be estimated as $|\boldsymbol{v}_{\rm rel}| \sim \tau_{\rm p} |(\boldsymbol{u} \cdot \boldsymbol{\nabla}) \boldsymbol{u}| \sim \tau_{\rm p} u_\eta^2/\eta$. Thus the collisions in clusters may be essential for

$$n_{\rm cl} \sim a^{-3} (\eta/a)(\rho/3\rho_{\rm p}) \,, \quad \ell_{\rm s} \sim a (3 a \rho_{\rm p} / \eta \rho)^{1/3} \,, \tag{4.2}$$

where $\ell_{\rm s}$ is a mean separation of particles in the cluster. For the above parameters ($a = 30\mu$m) $n_{\rm cl} \sim 3 \times 10^5$cm^{-3}, $\ell_{\rm s} \sim 5\,a \approx 150\mu$m and $N_{\rm cl} \sim 300$. Note that the mean number density of droplets in clouds $\bar n$ is about $10^2 - 10^3$cm^{-3}. Therefore the *clustering instability of droplets in the clouds increases their concentrations in the clusters by the orders of magnitude.*

In all our analysis we have neglected the effect of sedimentation of particles in gravity field which is essential for particles of the radius $a > 100\mu$m. Taking $\ell_{\rm cl} \simeq \eta$ we assumed implicitly that $\tau_{\rm p} < \tau_\eta$. This is valid (for the atmospheric conditions) if $a \leq 60\mu$m. Otherwise the cluster size can be estimated as $\ell_{\rm cl} \simeq \eta (\tau_{\rm p}/\tau_\eta)^{3/2}$.

Our estimates support the conjecture that *the clustering instability serves as a preliminary stage for a coagulation of water droplets in clouds leading to a rain formation.*

V. DISCUSSION

In this study we investigated the clustering instability of the spatial distribution of inertial particles advected by a turbulent velocity field. The instability results in formation of clusters, *i.e.,* small-scale inhomogeneities of aerosols and droplets. The clustering instability is caused by a combined effect of the particle inertia and finite correlation time of the velocity field. The finite correlation time of the turbulent velocity field causes the compressibility of the field of Lagrangian trajectories. The latter implies that the number of particles flowing into a small control volume in a Lagrangian frame does not equal to the number of particles flowing out from this control volume during a correlation time. This can result in the depletion of turbulent diffusion.

The role of the compressibility of the velocity field is as follows. Divergence of the velocity field of the inertial particles $\operatorname{div} \boldsymbol{v} = \tau_p \Delta P/\rho$. The inertia of particles results in that particles inside the turbulent eddies are carried out to the boundary regions between the eddies by inertial forces (i.e., regions with low vorticity and high strain rate). For a small molecular diffusivity $\operatorname{div} \boldsymbol{v} \propto -dn/dt$ [see Eq. (2.1)]. Therefore, $dn/dt \propto -\tau_p \Delta P/\rho$. Thus there is accumulation of inertial particles (i.e., $dn/dt > 0$) in regions with $\Delta P < 0$. Similarly, there is an outflow of inertial particles from the regions with $\Delta P > 0$. This mechanism acts in a wide range of scales of a turbulent fluid flow. Turbulent diffusion results in relaxation of fluctuations of particles concentration in large scales. However, in small scales where turbulent diffusion is small, the relaxation of fluctuations of particle concentration is very weak. Therefore the fluctuations of particle concentration are localized in the small scales.

This phenomenon is considered for the case when density of fluid is much less than the material density ρ_p of particles ($\rho \ll \rho_p$). When $\rho \geq \rho_p$ the results coincide with those obtained for the case $\rho \ll \rho_p$ except for the transformation $\tau_p \to \beta_* \tau_p$, where

$$\beta_* = 2\biggl(1 + \frac{\rho}{\rho_p}\biggr)\biggl(\frac{\rho_p - \rho}{2\rho_p + \rho}\biggr) \, .$$

For $\rho \geq \rho_p$ the value $dn/dt \propto -\beta_* \tau_p \Delta P/\rho$. Thus there is accumulation of inertial particles (i.e., $dn/dt > 0$) in regions with the minimum pressure of a turbulent fluid since $\beta_* < 0$. In the case $\rho \geq \rho_p$ we used the equation of motion of particles in fluid flow which takes into account contributions due to the pressure gradient in the fluid surrounding the particle (caused by acceleration of the fluid) and the virtual ("added") mass of the particles relative to the ambient fluid [39].

The exponential growth of the second moment of a number density of inertial particles due to the small-

scale instability can be saturated by the nonlinear effects (see Section IV). The excitation of the second moment of a number density of particles requires two kinds of compressibilities: compressibility of the velocity field and compressibility of the field of Lagrangian trajectories, which is caused by a finite correlation time of a random velocity field. Remarkably, the compressibility of the field of Lagrangian trajectories determines the coefficient of turbulent diffusion (i.e. the coefficient $D_{\alpha\beta}$ near the second-order spatial derivative of the second moment of a number density of inertial particles in Eq. (3.2). The compressibility of the field of Lagrangian trajectories causes depletion of turbulent diffusion in small scales even for $\sigma_v = 0$. On the other hand, the compressibility of the velocity field determines a coefficient B(r) near the second moment of a number density of inertial particles in Eq. (3.2). This term is responsible for the exponential growth of the second moment of a number density of particles.

Summary:

• We showed that the physical reason for the *clustering instability* in spatial distribution of particles in turbulent flows is a combined effect of the inertia of particles leading to a compressibility of the particle velocity field $\boldsymbol{v}(t,\boldsymbol{r})$ and a finite velocity correlation time.

• The clustering instability can result in a *strong clustering* whereby a finite fraction of particles is accumulated in the clusters and a *weak clustering* when a finite fraction of particle collisions occurs in the clusters.

• The crucial parameter for the clustering instability is a radius of the particles a. The instability criterion is $a > a_{\rm cr} \approx a_*$ for which $\langle(\mathrm{div}\,\boldsymbol{v})^2\rangle = \langle|\mathrm{rot}\,\boldsymbol{v}|^2\rangle$. For the droplets in the atmosphere $a_* \simeq 30\mu$m. The growth rate of the clustering instability $\gamma_{\rm cl} \sim \tau_\eta^{-1}(a/a_*)^4$, where τ_η is the turnover time in the viscous scales of turbulence.

• We introduced a new concept of compressibility of the turbulent diffusion tensor caused by a finite correlation time of an incompressible velocity field. For this model of the velocity field, the field of Lagrangian trajectories is not divergence-free.

• We suggested a mechanism of saturation of the clustering instability - *particle collisions in the clusters*. An evaluated nonlinear level of the saturation of the droplets number density in clouds exceeds by the orders of magnitude their mean number density.

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APPENDIX A: Basic equations in the model with a random renewal time

In this Appendix we derive Eq. (A20) for the simultaneous second-order correlation function $\Phi(t,\boldsymbol{r})$ which serves as a basis for further analysis in Appendixes B and C under some simplifying model assumptions about the statistics of the velocity field.

1. Exact solution of dynamical equations for a given velocity field

a. *Simple case: no molecular diffusion*

Consider first Eq. (2.1) for the number density of particles $n(t,\boldsymbol{r})$ in the case $D=0$:

$$\frac{\partial n(t,\boldsymbol{r})}{\partial t} + \boldsymbol{\nabla}\cdot[n(t,\boldsymbol{r})\boldsymbol{v}(t,\boldsymbol{r})] = 0\,, \tag{A1}$$

when all particles are transported only by advection. Solution of Eq. (A1) with the initial condition $n(s,\boldsymbol{r})$ is given by

$$n(t,\boldsymbol{r}) \;=\; G(t,\boldsymbol{r})\, n[s,\boldsymbol{\xi}_{\rm L}(t,\boldsymbol{r}|s)]\,, \tag{A2}$$

where $\boldsymbol{\xi}_{\rm L}(t,\boldsymbol{r}|s)$ is the Lagrangian trajectory of the particle which is located at coordinate $\boldsymbol{r}$ at time t. Here we label the particles at *present* moment of time t and consider a *current* time $s<t$ as moments in the past. This differs from a usual approach, see Eqs. (2.4), when particles are labelled at the *initial* time t_0, and a current time $t>t_0$. Therefore in the equations below it is more convenient to redefine Lagrangian displacement $\boldsymbol{\rho}_{\rm L}(t,r|s) \to \tilde{\boldsymbol{\rho}}_{\rm L}(t,r|s) = -\boldsymbol{\rho}_{\rm L}(t,r|s)$. Now Eqs. (2.4) can be written as

$$\tilde{\boldsymbol{\rho}}_{\rm L}(t,\boldsymbol{r}|s) \;=\; \int_s^t \boldsymbol{v}[\tau,\boldsymbol{\xi}_{\rm L}(t,\boldsymbol{r}|\tau)]\,\mathrm{d}\tau\,, \tag{A3}$$

$$\boldsymbol{\xi}_{\rm L}(t,\boldsymbol{r}|s) \;\equiv\; \boldsymbol{r} - \tilde{\boldsymbol{\rho}}_{\rm L}(t,\boldsymbol{r}|s)\;. \tag{A4}$$

The Green function is the functional of $\boldsymbol{\xi}_{\rm L}(t,\boldsymbol{r}|s)$:

$$G(t,\boldsymbol{r},s) \;=\; \exp\Big\{-\int_s^t b[\tau,\boldsymbol{\xi}_{\rm L}(t,\boldsymbol{r}|\tau)]\,d\tau\Big\}\,,$$

$$b(t,\boldsymbol{r}) \;\equiv\; \boldsymbol{\nabla}\cdot\boldsymbol{v}(t,\boldsymbol{r})\,, \tag{A5}$$

Introduce the *shift operator*

$$\exp[-\tilde{\boldsymbol{\rho}}_{\rm L}\cdot\boldsymbol{\nabla}\,] = 1 - \tilde{\boldsymbol{\rho}}_{\rm L}\cdot\boldsymbol{\nabla} + \frac{1}{2!}[-\tilde{\boldsymbol{\rho}}_{\rm L}\cdot\boldsymbol{\nabla}]^2 - \ldots \tag{A6}$$

which acts as follows:

$$\exp[-\tilde{\boldsymbol{\rho}}_{\rm L}\cdot\boldsymbol{\nabla}\,]n(t,\boldsymbol{r}) = n(t,\boldsymbol{r}-\tilde{\boldsymbol{\rho}}_{\rm L})\;. \tag{A7}$$

One can validate relation (A7) by Taylor series expansion of the function $n(t, \boldsymbol{r} - \tilde{\boldsymbol{\rho}}_{\rm L})$. Now Eq. (A2) can be rewritten as follows:

$$n(t, \boldsymbol{r}) = G(t, \boldsymbol{r}, s)\, \exp[-\tilde{\boldsymbol{\rho}}_{\rm L}(t, \boldsymbol{r}|s) \cdot \boldsymbol{\nabla}\,] n(s, \boldsymbol{r})\ . \quad \text{(A8)}$$

b. Molecular diffusion as a Wiener process

Consider now the full Eq. (2.1) with $D \neq 0$ whereby particles are transported by both, fluid advection and molecular diffusion. It was found by Wiener (see, e.g., [33]) that Brownian motion (molecular diffusion) can be described by the *Wiener* random process $\boldsymbol{w}(t)$ with the following properties:

$$\langle \boldsymbol{w}(t) \rangle_{\boldsymbol{w}} = 0\,, \quad \langle w_i(t+\tau) w_j(t) \rangle_{\boldsymbol{w}} = \tau \delta_{ij}\ . \quad \text{(A9)}$$

Here $\langle \dots \rangle_{\boldsymbol{w}}$ denotes the mathematical expectation over the statistics of the Wiener process. Introduce the Wiener trajectory $\boldsymbol{\xi}_{\rm W}(t, \boldsymbol{r}|s)$ (which usually is called the *Wiener path*) and the Wiener displacement $\boldsymbol{\rho}_{\rm W}(t, \boldsymbol{r}|s)$ as follows:

$$\begin{aligned} \boldsymbol{\xi}_{\boldsymbol{w}}(t, \boldsymbol{r}|s) &\equiv \boldsymbol{r} - \boldsymbol{\rho}_{\rm W}(t, \boldsymbol{r}|s)\,, && \text{(A10)} \\ \boldsymbol{\rho}_{\rm W}(t, \boldsymbol{r}|s) &= \int_s^t \boldsymbol{v}[\tau, \boldsymbol{\xi}_{\boldsymbol{w}}(t, \boldsymbol{r}|\tau)]\, {\rm d}\tau + \sqrt{2D}\boldsymbol{w}(t-s)\ . \end{aligned}$$

Comparison of this formula with Eqs. (A3) shows that in the limit $D \to 0$, $\boldsymbol{\xi}_{\rm W}(t, \boldsymbol{r}|s) \to \boldsymbol{\xi}_{\rm L}(t, \boldsymbol{r}|s)$ and $\boldsymbol{\rho}_{\rm W}(t, \boldsymbol{r}|s) \to \boldsymbol{\rho}_{\rm L}(t, \boldsymbol{r}|s)$.

In Refs. [35] it was shown that solution of Eq. (2.1) (with $D \neq 0$) can be written as solution (A8) of Eq. (A1) (with $D = 0$) by replacement $\tilde{\boldsymbol{\rho}}_{\rm L}(t, \boldsymbol{r}|s) \to \tilde{\boldsymbol{\rho}}_{\rm W}(t, \boldsymbol{r}|s)$ and then averaging over the statistics of the Wiener processes (A9):

$$n(t, \boldsymbol{r}) = \langle G(t, \boldsymbol{r}, s)\, \exp[-\boldsymbol{\rho}_{\rm W}(t, \boldsymbol{r}|s) \cdot \boldsymbol{\nabla}\,] n(s, \boldsymbol{r}) \rangle_{\boldsymbol{w}}\ . \quad \text{(A11)}$$

2. Two-step averaging over velocity statistics

a. Model of a random velocity field

Note that Eq. (A11) is a solution of Eq. (2.1) at a *given* realization of the random velocity field. Our next goal is to determine the simultaneous correlation functions

$$\begin{aligned} \bar{n}(t) &= \langle\!\langle n(t, \boldsymbol{r}) \rangle\!\rangle_{\boldsymbol{v}}\,, && \text{(A12)} \\ \Phi(t, \boldsymbol{r}_2 - \boldsymbol{r}_1) &= \langle\!\langle n(t, \boldsymbol{r}_1) n(t, \boldsymbol{r}_2) \rangle\!\rangle_{\boldsymbol{v}} - \bar{n}^2(t)\,, \end{aligned}$$

averaged over the stationary, space homogeneous statistics of turbulent velocity field, where $\langle\!\langle \dots \rangle\!\rangle_{\boldsymbol{v}}$ denotes this averaging. Since the initial distribution $n(t_0, \boldsymbol{r})$ is assumed to be homogeneous in space, $\bar{n}(t)$ is independent of spatial coordinate, and $\Phi(t, \boldsymbol{r}_2 - \boldsymbol{r}_1)$ depends only on the difference $\boldsymbol{r}_2 - \boldsymbol{r}_1$.

In order to simplify the averaging procedure (A12) we consider a model of random velocity field which fully looses memory at some instants of renewal τ_j. For t_1 and t_2 inside a renewal interval $[\tau_j < t_1, t_2 < \tau_{j+1}]$ the velocity pair correlation function is defined as

$$\mathcal{F}^{\alpha\beta}(t_2 - t_1, \boldsymbol{r}_2 - \boldsymbol{r}_1) \equiv \langle v_\alpha(t_1, \boldsymbol{r}_1) v_\beta(t_2, \boldsymbol{r}_2) \rangle_{\boldsymbol{v}}\,, \quad \text{(A13)}$$

where $\langle \dots \rangle_{\boldsymbol{v}}$ denotes averaging over "intrinsic statistics" of the velocity field. In our model the velocity fields before and after renewals are statistically independent. The interval between the renewal instants τ_j may be the same or randomly distributed, say with the Poisson statistics. In the latter case the full averaging $\langle\!\langle \dots \rangle\!\rangle_{\boldsymbol{v}}$ may be considered as a two-stage process. First one calculates $\langle \dots \rangle_{\boldsymbol{v}}$ and then averages over the statistics of the renewal time $\tau_{\rm ren}$, which is denoted as $\langle \dots \rangle_{\rm ren}$:

$$\langle\!\langle \dots \rangle\!\rangle_{\boldsymbol{v}} \equiv \langle\ \langle \dots \rangle_{\boldsymbol{v}} \rangle_{\rm ren}\ . \quad \text{(A14)}$$

For the Poisson statistics of τ_j

$$\begin{aligned} & F^{\alpha\beta}(t_2 - t_1, \boldsymbol{r}_2 - \boldsymbol{r}_1) \equiv \langle\!\langle v_\alpha(t_1, \boldsymbol{r}_1) v_\beta(t_2, \boldsymbol{r}_2) \rangle\!\rangle_{\boldsymbol{v}} \\ &= \mathcal{F}^{\alpha\beta}(t_2 - t_1, \boldsymbol{r}_2 - \boldsymbol{r}_1) \exp(-|t_2 - t_1|/\bar{\tau}_{\rm ren})\,, \quad \text{(A15)} \end{aligned}$$

where $\bar{\tau}_{\rm ren}$ is a mean renewal time. It would be useful to define the correlation time of the function $\mathcal{F}^{\alpha\beta}$ as follows

$$\tau_v(R) = \int \mathcal{F}^{\alpha\beta}(\tau, \boldsymbol{R})\, d\tau / \mathcal{F}^{\alpha\beta}(0, \boldsymbol{R})\ . \quad \text{(A16)}$$

Certainly this model of the random velocity field cannot be considered as universal. However, it reproduces important features of some flows (see, e.g., Ref. [40]).

b. Averaging procedure

Our model involves three random processes:

1. The Wiener random process which describes Brownian (molecular) diffusion.

2. Poisson process for a random renewal time.

3. The random velocity field between the renewals.

Eq. (A11) presents $n(t, \boldsymbol{r})$ after the first step, i.e., it describes the number density at a *given* realization of a velocity field. Using Eq. (A11) we obtain

$$\begin{aligned} & n(t, \boldsymbol{r}_1) n(t, \boldsymbol{r}_2) = \langle G(\boldsymbol{r}_1) G(\boldsymbol{r}_2)\, \exp[\boldsymbol{\xi}'(\boldsymbol{r}_1) \cdot \boldsymbol{\nabla}_1 \\ & + \boldsymbol{\xi}'(\boldsymbol{r}_2) \cdot \boldsymbol{\nabla}_2] n(s, \boldsymbol{r}_1) n(s, \boldsymbol{r}_2) \rangle_{\boldsymbol{ww}}\,, \quad \text{(A17)} \end{aligned}$$

where $\boldsymbol{\nabla}_1 = \partial/\partial \boldsymbol{r}_1$ and $\boldsymbol{\nabla}_2 = \partial/\partial \boldsymbol{r}_2$ and $\langle \rangle_{\boldsymbol{ww}}$ denotes averaging over two independent Wiener processes determining two Wiener paths. Hereafter for simplicity we use the following notations: $G(\boldsymbol{r}) \equiv G(t, \boldsymbol{r}, s)$ and $\boldsymbol{\xi}'(\boldsymbol{r}) \equiv \boldsymbol{\xi}'(t, \boldsymbol{r}|s)$.

Now we average Eq. (A17) over a random velocity field for a given realization of a Poisson process:

$$\tilde{\Phi}(t,\boldsymbol{r}_2-\boldsymbol{r}_1) = \langle n(t,\boldsymbol{r}_1)n(t,\boldsymbol{r}_2)\rangle_{\boldsymbol{v}} - (\bar{n})^2 \qquad \text{(A18)}$$
$$= \langle\langle G(\boldsymbol{r}_1)G(\boldsymbol{r}_2)\, \exp[\boldsymbol{\xi}'(\boldsymbol{r}_1)\cdot\boldsymbol{\nabla}_1 + \boldsymbol{\xi}'(\boldsymbol{r}_2)\cdot\boldsymbol{\nabla}_2]\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}}$$
$$\times\tilde{\Phi}(t_0,\boldsymbol{r}_1-\boldsymbol{r}_2)\} \ .$$

Here the time t_0 is the last renewal time before time t and $t' = t - t_0$ is a random variable. Thus, averaging of the functions

$$G(\boldsymbol{r}_1)G(\boldsymbol{r}_2)\, \exp[\boldsymbol{\xi}'(\boldsymbol{r}_1)\cdot\boldsymbol{\nabla}_1 + \boldsymbol{\xi}'(\boldsymbol{r}_2)\cdot\boldsymbol{\nabla}_2]\, , \Phi(t_0,\boldsymbol{r}_1-\boldsymbol{r}_2)$$

is decoupled into two time intervals because the first function is determined by the velocity field after the renewal while the second function $\Phi(t_0,\boldsymbol{r}_1-\boldsymbol{r}_2)$ is determined by the velocity field before renewal. Now we take into account that for the Poisson process any instant can be chosen as the initial instant. We average Eq. (A18) over the random renewal time. The probability density $p(t')$ for a random renewal time is given by

$$p(t') = \bar{\tau}_{\rm ren}^{-1} \exp(-t'/\bar{\tau}_{\rm ren}) \ . \qquad \text{(A19)}$$

Thus the resulting averaged equation for "fully" averaged correlation function $\Phi(t,\boldsymbol{R}) = \langle\tilde{\Phi}(t,\boldsymbol{R})\rangle_{\rm ren}$, defined by Eq. (A12), assumes the following form:

$$\Phi(t,\boldsymbol{R}) = \bar{\tau}_{\rm ren}^{-1}\int_0^t \hat{P}(\tau,\boldsymbol{R})\Phi(t-\tau,\boldsymbol{R})\exp(-\tau/\bar{\tau}_{\rm ren})\, d\tau$$
$$+ \exp(-t/\bar{\tau}_{\rm ren})\hat{P}(t,\boldsymbol{R})\Phi_0(\boldsymbol{R}) \, . \qquad \text{(A20)}$$

The first term in Eq. (A20) describes the case when there is at least one renewal of the velocity field during the time t (i.e., the Poisson event), whereas the second term describes the case when there is no renewal during the time t. Here $\Phi_0(\boldsymbol{R}) = \Phi(t=0,\boldsymbol{R})$ and

$$\hat{P}(t,\boldsymbol{R}) = \langle\langle G(\boldsymbol{r}_1)G(\boldsymbol{r}_2) \qquad \text{(A21)}$$
$$\times \exp[\boldsymbol{\xi}'(\boldsymbol{r}_1)\cdot\boldsymbol{\nabla}_1 + \boldsymbol{\xi}'(\boldsymbol{r}_2)\cdot\boldsymbol{\nabla}_2]\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}}$$
$$= \exp\langle\langle g(\boldsymbol{r}_1) + g(\boldsymbol{r}_2) + \boldsymbol{\xi}'(\boldsymbol{r}_1)\cdot\boldsymbol{\nabla}_1 + \boldsymbol{\xi}'(\boldsymbol{r}_2)\cdot\boldsymbol{\nabla}_2\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}} \, ,$$

where $G(\boldsymbol{r}) = \exp[g(\boldsymbol{r})]$. Equation (A20) is simplified in Appendixes B and C under the additional assumptions about the velocity field statistics.

APPENDIX B: VELOCITY FIELD WITH GAUSSIAN LAGRANGIAN TRAJECTORIES

Consider a model of a random velocity field where Lagrangian trajectories, i.e., *the integrals* $\int \boldsymbol{v}(\mu,\boldsymbol{\xi})\, d\mu$ *and* $\int b(\mu,\boldsymbol{\xi})\, d\mu$ have Gaussian statistics. Using an identity $\langle\exp(a\eta)\rangle_\eta = \exp(\frac{1}{2}a^2)$ in Eq. (A21) we obtain

$$\hat{P}(\mu,\boldsymbol{R}) \ = \ \exp[\mu\hat{\mathcal{L}}] \ , \qquad \text{(B1)}$$

where

$$\begin{aligned}
\hat{\mathcal{L}} \ &= \ B(\boldsymbol{R}) + 2U_\alpha(\boldsymbol{R})\nabla_\alpha + \hat{D}_{\alpha\beta}(\boldsymbol{R})\nabla_\alpha\nabla_\beta \, , \\
\mu B(\boldsymbol{R}) \ &= \ \langle\langle g(\boldsymbol{r}_1)g(\boldsymbol{r}_2)\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}} \ , \qquad \text{(B2)} \\
\mu U_\alpha(\boldsymbol{R}) \ &= \ -\langle\langle \xi'_\alpha(\boldsymbol{r}_1)g(\boldsymbol{r}_2)\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}} \ , \\
\hat{D}_{\alpha\beta}(\boldsymbol{R}) \ &= \ D_{\alpha\beta}(0) - D_{\alpha\beta}(\boldsymbol{R}) \ , \\
\mu D_{\alpha\beta}(\boldsymbol{R}) \ &= \ \langle\langle \xi'_\alpha(\boldsymbol{r}_1)\xi'_\beta(\boldsymbol{r}_2)\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}} \ .
\end{aligned}$$

Here η is a Gaussian random variable with zero mean value and unit variance and $\langle\langle G(\boldsymbol{r})\rangle_{\boldsymbol{w}}\rangle_{\boldsymbol{v}} = 1$. The latter yields

$$\langle\langle g\rangle_{\boldsymbol{w}}\rangle_{\boldsymbol{v}} = -\frac{1}{2}\langle\langle \tilde{g}^2\rangle_{\boldsymbol{w}}\rangle_{\boldsymbol{v}} \, , \quad g = \langle\langle g\rangle_{\boldsymbol{w}}\rangle_{\boldsymbol{v}} + \tilde{g} \, ,$$

where $\langle\langle \tilde{g}\rangle_{\boldsymbol{w}}\rangle_{\boldsymbol{v}} = 0$. When correlation time $\tau_v(R)$, Eq. (A16), is much less then the current time t and $\bar{\tau}_{\rm ren}$, these correlation functions are given by

$$B(\boldsymbol{R}) = 2\int_0^\infty \langle\langle b[0,\boldsymbol{\xi}(\boldsymbol{r}_1)]b[\mu',\boldsymbol{\xi}(\boldsymbol{r}_2)]\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}}\, d\mu', \qquad \text{(B3)}$$
$$U_\alpha(\boldsymbol{R}) = -2\int_0^\infty \langle\langle v_\alpha[0,\boldsymbol{\xi}(\boldsymbol{r}_1)]b[\mu',\boldsymbol{\xi}(\boldsymbol{r}_2)]\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}}\, d\mu' \ ,$$
$$D_{\alpha\beta}(\boldsymbol{R}) = 2\int_0^\infty \langle\langle v_\alpha[0,\boldsymbol{\xi}(\boldsymbol{r}_1)]v_\beta[\mu',\boldsymbol{\xi}(\boldsymbol{r}_2)]\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}}\, d\mu' \ ,$$

where we used an identity

$$\langle\langle \int_0^\mu a_\alpha(\mu',\boldsymbol{r}_1)\, d\mu' \int_0^\mu c_\beta(\mu'',\boldsymbol{r}_2)\, d\mu''\rangle_{\boldsymbol{w}}\rangle_{\boldsymbol{v}}$$
$$\simeq 2\mu\int_0^\infty \langle\langle a_\alpha(0,\boldsymbol{r}_1)c_\beta(\mu',\boldsymbol{r}_2)\rangle_{\boldsymbol{w}}\rangle_{\boldsymbol{v}}\, d\mu' \ .$$

Eq. (B1) allows to rewrite Eq. (A20) as

$$\Phi(t,\boldsymbol{R}) \ = \ \frac{1}{\bar{\tau}_{\rm ren}}\left[\int_0^t \exp(\mu\hat{\mathcal{L}}_1)\, d\mu\right]\Phi(t,\boldsymbol{R})$$
$$+ \exp(t\hat{\mathcal{L}}_1)\Phi(t,\boldsymbol{R}) \, , \qquad \text{(B4)}$$

where

$$\hat{\mathcal{L}}_1 = \hat{\mathcal{L}} - \frac{\partial}{\partial t} - \frac{1}{\bar{\tau}_{\rm ren}} \ . \qquad \text{(B5)}$$

To derive Eq. (B4) we used the following identity

$$\Phi(t-\mu,\boldsymbol{R}) = \exp\biggl(-\mu\frac{\partial}{\partial t}\biggr)\Phi(t,\boldsymbol{R}) \ , \qquad \text{(B6)}$$

which follows from the Taylor expansion

$$f(t+\tau) = \sum_{m=1}^\infty \biggl(\tau\frac{\partial}{\partial t}\biggr)^m \frac{f(t)}{m!} = \exp\biggl(\tau\frac{\partial}{\partial t}\biggr)f(t) \ . \qquad \text{(B7)}$$

In particular,

$$\Phi_0(\boldsymbol{R}) = \Phi(t-t,\boldsymbol{R}) = \exp\left(-t\frac{\partial}{\partial t}\right)\Phi(t,\boldsymbol{R}) \ .$$

Evaluating the integral in Eq. (B4) we obtain

$$\left[\exp(t\hat{\mathcal{L}}_1) - 1\right]\left(\hat{\mathcal{L}}_1 + \bar{\tau}_{\rm ren}^{-1}\right)\Phi(t,\boldsymbol{R}) = 0 \ . \qquad \text{(B8)}$$

Here we used the commutativity relation

$$\hat{\mathcal{L}}_1 \exp(t\hat{\mathcal{L}}_1) = \exp(t\hat{\mathcal{L}}_1)\,\hat{\mathcal{L}}_1 \ .$$

Thus, finally

$$\frac{\partial\Phi}{\partial t} = \Big[B(\boldsymbol{R}) + 2\mathbf{U}(\boldsymbol{R})\cdot\boldsymbol{\nabla} + \hat{D}_{\alpha\beta}(\boldsymbol{R})\nabla_\alpha\nabla_\beta\Big]\Phi(t,\boldsymbol{R}) \,, \qquad \text{(B9)}$$

Note that in the limit $\bar{\tau}_{\rm ren}\to\infty$, Eq. (B9) describes the evolution of $\Phi(t,\boldsymbol{R})$ in the model of the random velocity field without renewals.

APPENDIX C: GAUSSIAN VELOCITY FIELD WITH A SMALL YET FINITE CORRELATION TIME

Here we consider a random Gaussian velocity field with a small $\bar{\tau}_{\rm ren}$. Using Eq. (B6) we rewrite Eq. (A20) in the form

$$\left\{\frac{1}{\bar{\tau}_{\rm ren}}\int\limits_0^t \hat{P}(\tau,\boldsymbol{R})\exp\left(-\frac{\tau}{\bar{\tau}_{\rm ren}}\hat{M}\right)d\tau - 1\right\}\Phi(t,\boldsymbol{R}) = 0 \,, \qquad \text{(C1)}$$

where $\hat{M} = 1 + \bar{\tau}_{\rm ren}(\partial/\partial t)$ and we neglected the last term in Eq. (A20) for small τ. Expanding the function $\hat{P}(\tau,\boldsymbol{R})$ in Taylor series in the vicinity of $\tau=0$ we obtain

$$\Big\{\sum_{k=0}^{\infty}\bar{\tau}_{\rm ren}^{k}\Big[\frac{\partial^k\hat{P}(\tau,\boldsymbol{R})}{\partial\tau^k}\Big]_{\tau=0}\hat{M}^{-(k+1)} - 1\Big\}\Phi(t,\boldsymbol{R}) = 0 \,, \qquad \text{(C2)}$$

where we used that

$$\int\limits_0^t \tau^k\exp\left(-\frac{\tau}{\bar{\tau}_{\rm ren}}\hat{M}\right)d\tau = k!\,\bar{\tau}_{\rm ren}^{k+1}\hat{M}^{-(k+1)} \ .$$

Neglecting the terms $\sim O(\bar{\tau}_{\rm ren}^{5})$ in Eq. (C2) we obtain

$$\hat{M}^2\frac{\partial\Phi(t,\boldsymbol{R})}{\partial t} = \bar{\tau}_{\rm ren}\Bigg[\left(\frac{\partial^2\hat{P}(\tau,\boldsymbol{R})}{\partial\tau^2}\right)_{\tau=0} + \bar{\tau}_{\rm ren}^2\left(\frac{\partial^4\hat{P}(\tau,\boldsymbol{R})}{\partial\tau^4}\right)_{\tau=0}\Bigg]\Phi(t,\boldsymbol{R}) \,, \qquad \text{(C3)}$$

where we used that the expansion of the operator $\hat{P}(\tau,\boldsymbol{R})$ into Taylor series (for small τ) for a random Gaussian velocity field has only even powers of τ. Thus, the equation for the correlation function $\Phi(t,\boldsymbol{R})$ is given by

$$\hat{M}^2\frac{\partial\Phi(t,\boldsymbol{R})}{\partial t} = [B(\boldsymbol{R}) + 2\boldsymbol{U}(\boldsymbol{R})\cdot\boldsymbol{\nabla} + \hat{D}_{\alpha\beta}(\boldsymbol{R})\nabla_\alpha\nabla_\beta]\Phi \,, \qquad \text{(C4)}$$

where

$$\hat{D}_{\alpha\beta}(\boldsymbol{R}) = \frac{\bar{\tau}_{\rm ren}}{2}\langle\langle\tilde{\xi}_\alpha\tilde{\xi}_\beta G(\boldsymbol{r}_1)G(\boldsymbol{r}_2)\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}} \,, \qquad \text{(C5)}$$

$$U_\alpha(\boldsymbol{R}) = -\frac{1}{\bar{\tau}_{\rm ren}}\langle\langle g(\boldsymbol{r}_2)\xi_\alpha^*(\boldsymbol{r}_1)\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}} + \frac{1}{2\bar{\tau}_{\rm ren}}\langle\langle g(\boldsymbol{r}_1)g(\boldsymbol{r}_2)\tilde{\xi}_\alpha\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}} \,, \qquad \text{(C6)}$$

$$B(\boldsymbol{R}) = \frac{1}{\bar{\tau}_{\rm ren}}\langle\langle g(\boldsymbol{r}_1)g(\boldsymbol{r}_1)\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}} \,, \qquad \text{(C7)}$$

Here for the homogeneous turbulent velocity field [21]:

$$\tilde{\boldsymbol{\xi}} = \boldsymbol{\xi}'(\boldsymbol{r}_2) - \boldsymbol{\xi}'(\boldsymbol{r}_1) \,, \qquad \boldsymbol{\nabla} = \partial/\partial\boldsymbol{R} \,, \qquad G = \bar{G} + g \,, \qquad \langle\langle g\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}} = 0 \,, \qquad \bar{G} = \langle\langle G\rangle_{\boldsymbol{ww}}\rangle_{\boldsymbol{v}} = 1 \ .$$

Using the expansion of $\boldsymbol{\xi}(\bar{\tau}_{\rm ren},\boldsymbol{r})$ and $g[\bar{\tau}_{\rm ren},\boldsymbol{\xi}(\boldsymbol{r})]$ into Taylor series of a small time $\bar{\tau}_{\rm ren}$ after the lengthly algebra we obtain

$$\hat{D}_{\alpha\beta}(\boldsymbol{R}) = 2D\delta_{\alpha\beta} + 2\bar{\tau}_{\rm ren}[\tilde{f}_{\alpha\beta}(\boldsymbol{R}) + {\rm St}^2 Q_{\alpha\beta}(\boldsymbol{R})] \,, \qquad \text{(C8)}$$

$$Q_{\alpha\beta}(\boldsymbol{R}) = 3[(\nabla_\nu f_{\mu\beta})(\nabla_\mu f_{\alpha\nu}) - \tilde{f}_{\mu\nu}\nabla_\nu\nabla_\mu f_{\alpha\beta}] + 24A_\alpha A_\beta + 12(A_\mu\nabla_\mu f_{\alpha\beta} - \tilde{f}_{\alpha\beta}\nabla_\mu A_\mu) - 20\tilde{f}_{\alpha\mu}\nabla_\mu A_\beta \,, \tag{C9}$$

$$U_\alpha(\boldsymbol{R}) = -2\bar{\tau}_{\rm ren}\{A_\alpha - {\rm St}^2[(\nabla_\nu A_\mu)(\nabla_\mu f_{\alpha\nu}) + 10A_\mu\nabla_\mu A_\alpha + 12A_\alpha\nabla_\mu A_\mu]\} \,, \tag{C10}$$

$$B(\boldsymbol{R}) = -2\bar{\tau}_{\rm ren}\{\nabla_\mu A_\mu + {\rm St}^2[(\nabla_\nu A_\mu)(\nabla_\mu A_\nu) - 6(\nabla_\mu A_\mu)^2]\} \,, \tag{C11}$$

$$A_\alpha = \nabla_\beta f_{\alpha\beta} \,, \quad \tilde{f}_{\alpha\beta} = f_{\alpha\beta}(0) - f_{\alpha\beta}(\boldsymbol{R}) \,, \quad f_{\alpha\beta}(\boldsymbol{R}) = \langle v_\alpha(\boldsymbol{r}_1) v_\beta(\boldsymbol{r}_2)\rangle_{\boldsymbol{v}} \,,$$

and St$=\bar{\tau}_{\rm ren}/\tau_\eta$ is the Strouhal number. In these calculations we neglected small terms $\sim O({\rm St}^2 R^3 \nabla^3)$. Our analysis showed that the neglected small terms do not affect the growth rate of the clustering instability. In Eqs. (C9)-(C11) we assumed that the correlation function $f_{\alpha\beta}$ for homogeneous, isotropic and compressible velocity field is given by

$$f_{\alpha\beta}(\boldsymbol{R}) = \frac{u_\eta^2}{3}\Big[(F + F_c)\delta_{\alpha\beta} + \frac{RF'}{2}P_{\alpha\beta} + RF'_c R_{\alpha\beta}\Big] \,, \tag{C12}$$

(see [36]), and in scales $0 < R \ll 1$ incompressible $F(R)$ and compressible $F_c(R)$ components of the random velocity field are given by

$$F(R) = (1 - R^2)/(1 + \sigma_v) \,, \quad F_c(R) = \sigma_v F(R) \,,$$

in scales $R \geq 1$ the functions $F = F_c = 0$. Here R is measured in the units of η, $P_{\alpha\beta}(R) = \delta_{\alpha\beta} - R_{\alpha\beta}$, $R_{\alpha\beta} = R_\alpha R_\beta / R^2$ and $F' = dF/dR$. Turbulent diffusion tensor $D_{\alpha\beta}(R)$ is determined by the field of Lagrangian trajectories $\boldsymbol{\xi}$ [see Eq. (C5)]. Due to a finite correlation time of a random velocity the field of Lagrangian trajectories $\boldsymbol{\xi}$ is compressible even if the velocity field is incompressible ($\sigma_v = 0$). Indeed, for $\sigma_v = 0$ we obtain

$$\langle\langle(\boldsymbol{\nabla}\cdot\boldsymbol{\xi})^2\rangle_{\boldsymbol{w}}\rangle_{\boldsymbol{v}} = \frac{20}{3}{\rm St}^4 \,.$$

Using Eqs. (C8)-(C12) we calculate the functions $\hat{D}_{\alpha\beta}(\boldsymbol{R})$, $U_\alpha(\boldsymbol{R})$ and $B(\boldsymbol{R})$:

$$\hat{D}_{\alpha\beta}(\boldsymbol{R}) = [2D + R^2(a_3 + {\rm St}^2 b_6)]\delta_{\alpha\beta} + R^2(a_4 + {\rm St}^2 b_4)R_{\alpha\beta}, \tag{C13}$$

$$U_\alpha(\boldsymbol{R}) = -R_\alpha(a_5 + {\rm St}^2 b_5) \,, \tag{C14}$$

$$B = a_6 + {\rm St}^2 b_2 \,, \tag{C15}$$

where $b_2 = -\frac{51}{47}b_5$ and

$$a_5 = -\frac{20\sigma_v}{3(1+\sigma_v)} = -\frac{a_2}{3} \,, \quad a_3 = \frac{2\sigma_v + 4}{3(1+\sigma_v)} \,,$$

$$a_4 = \frac{4\sigma_v - 2}{3(1+\sigma_v)} \,, \quad b_5 = -\frac{2350}{27}\left(\frac{\sigma_v}{1+\sigma_v}\right)^2 \,,$$

$$b_6 = \frac{12 + 872\sigma_v + 433\sigma_v^2}{27(1+\sigma_v)^2} \,,$$

$$b_4 = \frac{2(12 - 203\sigma_v + 1033\sigma_v^2)}{27(1+\sigma_v)^2} \,.$$

We will show here that the combined effect of particles inertia ($\sigma_v \neq 0$) and finite correlation time (St $\neq 0$) results in the excitation of the clustering instability whereby under certain conditions there is a self-excitation of the second moment of a number density of inertial particles. This instability causes formation of small-scale inhomogeneities in spatial distribution of inertial particles.

The equation for the second-order correlation function for the number density of inertial particles reads

$$\hat{M}^2\frac{\partial\Phi(t,R)}{\partial t} = \frac{\Phi''}{m(R)} + \tilde{\lambda}(R)\Phi' + B\Phi \tag{C16}$$

[see Eqs. (C15)], where the time t is measured in units of t_η, and

$$\Phi' = \frac{\partial\Phi}{\partial R} \,, \quad \Phi'' = \frac{\partial^2\Phi}{\partial R^2} \,, \quad \frac{1}{m} = \frac{2(1+X^2)}{\rm Sc} \,,$$

$$\tilde{\lambda} = \frac{2[2 + X^2(1+2C)]}{R\,{\rm Sc}} \,, \quad C = \frac{a_1 + {\rm St}^2 b_1}{4\beta} \,,$$

$$\beta = \frac{a_2 + {\rm St}^2 b_3}{2} \,, \ X(R) = \sqrt{{\rm Sc}\beta}\,R, \ R = |\boldsymbol{r}_2 - \boldsymbol{r}_1| \,,$$

$$a_1 = \frac{2(19\sigma_v + 3)}{3(1+\sigma_v)} \,, \quad a_2 = \frac{2(3\sigma_v + 1)}{3(1+\sigma_v)} \,,$$

$$b_1 = -\frac{1}{27(1+\sigma_v)^2}(12 - 1278\sigma_v - 3067\sigma_v^2) \,,$$

$$b_3 = \frac{1}{27(1+\sigma_v)^2}(36 + 466\sigma_v + 2499\sigma_v^2) \,.$$

In order to obtain a solution of Eq. (C16) we use a separation of variables, i.e., we seek for a solution in the following form:

$$\Phi(t,R) = \hat{\Phi}(R)\exp(\gamma_2 t) \,,$$

whereby γ_2 is a free parameter which is determined using the boundary conditions

$$\hat{\Phi}(R=0) = 1 \,, \quad \hat{\Phi}(R\to\infty) = 0 \,.$$

Here γ_2 is measured in units of $1/t_\eta$. Since the function $\Phi(t,R)$ is the two-point correlation function, it has a global maximum at $R=0$ and therefore it satisfies the conditions:

$$\hat{\Phi}'(R=0) = 0 \,, \quad \hat{\Phi}''(R=0) < 0 \,,$$

$$\hat{\Phi}(R=0) > |\hat{\Phi}(R>0)| \,.$$

Then Eq. (C16) yields

$$\Gamma\hat{\Phi}(R) = \frac{1}{m(R)}\hat{\Phi}'' + \tilde{\lambda}(R)\hat{\Phi}' + B\hat{\Phi} \,, \tag{C17}$$

where $\Gamma = \gamma_2(1+\bar{\tau}_{\rm ren}\gamma_2)^2$. Equation (C17) has an exact solution for $0 \leq R < 1$:

$$\hat\Phi(X) = S(X) X (1+X^2)^{\mu/2} , \qquad \text{(C18)}$$
$$S(X) = {\rm Re}\{A_1 P_\zeta^\mu(iX) + A_2 Q_\zeta^\mu(iX)\} ,$$

and $P_\zeta^\mu(Z)$ and $Q_\zeta^\mu(Z)$ are the Legendre functions with imaginary argument

$$Z = iX \, , \; \mu = C - \frac{3}{2} \, , \; \zeta = -\frac{1}{2} \pm \sqrt{C^2 - \kappa} \, , \; \kappa = \frac{B - \Gamma}{2\beta} .$$

Solution of Eq. (C16) can be analyzed using asymptotics of the exact solution (C18). This asymptotic analysis is based on the separation of scales (see, e.g., [34, 36]). In particular, the solution of Eq. (C16) has different regions where the form of the functions $m(R)$ and $\tilde\lambda(R)$ are different. The functions $\hat\Phi(R)$ and $\hat\Phi'(R)$ in these different regions are matched at their boundaries in order to obtain continuous solution for the correlation function. Note that the most important part of the solution is localized in small scales (i.e., $R \ll 1$). Using the asymptotic analysis of the exact solution for $X \gg 1$ allowed us to obtain the necessary conditions of a small-scale instability of the second moment of a number density of inertial particles. The results obtained by this asymptotic analysis are presented below.

The solution (C18) has the following asymptotics: for $X \ll 1$ (i.e., in the scales $0 \leq R \ll 1/\sqrt{\rm Sc}$) the solution for the second moment $\hat\Phi$ is given by

$$\hat\Phi(X) = \{1 - (\kappa/6)[X^2 + O(X^4)]\} . \qquad \text{(C19)}$$

For $X \gg 1$ (i.e., in the scales $1/\sqrt{\rm Sc} \ll R < 1$) the function $\hat\Phi$ is given by

$$\hat\Phi(X) = {\rm Re}\{A X^{-C \pm \sqrt{C^2-\kappa}}\}. \qquad \text{(C20)}$$

When $C^2 - \kappa < 0$ the second-order correlation function for a number density of inertial particles $\hat\Phi$ is given by

$$\hat\Phi(R) = A_3 R^{-C} \cos(\nu_I \ln R + \varphi) \, , \quad \nu_I = \sqrt{\kappa - C^2} \, ,$$

where $C > 0$ and φ is the argument of the complex constant A. For $R \geq 1$ the second-order correlation function for the number density of inertial particles is given by

$$\hat\Phi(R) = (A_4/R) \exp(-R \sqrt{3\Gamma/2}) \, , \qquad \text{(C21)}$$

where $\Gamma > 0$. Since the total number of particles in a closed volume is conserved, i.e., particles can only be redistributed in the volume,

$$\int_0^\infty R^2 \hat\Phi(R) \, dR = \hat\Phi(k=0) = 0 \; .$$

The latter yields $\varphi = -\pi/2$ for $\ln {\rm Sc} \gg 1$ and $\Gamma \ll 1$. When $C^2 - \kappa > 0$, the solution (C20) cannot be matched with solutions (C19) and (C21). Thus, the condition $C^2 - \kappa < 0$ is the necessary condition for the existence of the solution for the correlation function. The condition $C > 0$ provides the existence of the global maximum of the correlation function at $R = 0$.

Matching functions $\hat\Phi$ and $\hat\Phi'$ at the boundaries of the above-mentioned regions yields coefficients A_k and Γ. In particular, the eigenvalue Γ is given by Eq. (3.14).

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